ESTIMATION OF EXPOSURE OF PERSONS IN CALIFORNIA TO PESTICIDE PRODUCTS THAT CONTAIN TRIBUFOS (DEF)

BY

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ABSTRACT

Tribufos (DEF), S, S, S-tributyl phosphorotrithioate is currently registered in California as a cotton defoliant. There were a total of 16 illness and injury cases, mostly systemic in nature, associated with exposure to tribufos and tribufos in combination with other pesticides in California during 1982 to 1991. Formulated products of tribufos are strong dermal irritants. Tribufos is extensively (47.5 percent) absorbed through the skin of treated laboratory rats. Tribufos is absorbed rapidly and metabolized extensively in laboratory animals following oral administration. The absorbed daily dosages (ADD) for workers handling tribufos were estimated to range from 4 ug/kg/day for a ground applicator to 54 ug/kg/day for a ground mixer/loader. The ADD for workers involved in cotton harvesting ranged from 30 ug/kg/day for a module builder operator to 122 ug/kg/day for a tramper.

This report was prepared as part of the Department's risk characterization document for tribufos. Tribufos toxicity studies have shown cholinergic signs in exposed laboratory animals.

EXECUTIVE SUMMARY

Tribufos Humans Exposure Assessment
August 8, 1990
Revised June 20, 1995

Worker Health and Safety Branch
Department of Pesticide Regulation
California Environmental Protection Agency

PURPOSE

The Department of Pesticide Regulation is preparing a risk assessment document for tribufos because of possible adverse effects observed in the laboratory animals especially for cholinergic signs. This human exposure assessment will be incorporated into the risk characterization document for tribufos. This document reviews worker exposure studies under various work conditions in which workers handled tribufos or harvested tribufos-treated cotton fields. The document was revised to estimate Absorbed Daily Dosages (ADD) from the most recently available dermal absorption and worker exposure studies, based on the current worker protection statements on the product label.

BACKGROUND

Tribufos is an organophosphate pesticide that can cause cholinesterase inhibition in mammalian species. It is used for defoliation exclusively on cotton. Tribufos contains minute quantities of butyl mercaptan, a substance characterized by a strong skunk-like odor. DEF® 6 and Folex® 6EC, the formulated products of tribufos, are strong skin and eye irritants. When applied dermally to animals, tribufos is absorbed extensively. Tribufos is metabolized following oral administration and excreted rapidly in urine and feces in laboratory animals. Between 1982 and 1991, 16 cases of tribufos-associated illness and injury, mostly systemic, were reported to the Pesticide Illness Surveillance Program of Department of Pesticide Regulation.

Tribufos was placed into reevaluation in California on August 9, 1991 to determine the toxicity signal word and precautionary language on the product label which will sufficiently mitigate possible acute adverse effects. The reevaluation resulted in revising the precautionary statements and changing the toxicity signal word from 'Warning' to 'Danger'.

METHODS

This document reviewed studies in which workers were monitored for dermal and inhalation exposures to tribufos during product application and cotton harvest. It estimated ADD from the dermal and inhalation exposures for various work categories. The document also reviewed studies that monitored tribufos drift during applications and after the applications were completed.

FINDING and CONCLUSION

The ADD for workers handling tribufos were estimated to range from 4 ug/kg/day for a ground applicator to 54 ug/kg/day for a ground mixer/loader. The ADD for workers involved in cotton harvesting ranged from 30 ug/kg/day for a module builder operator to 122 ug/kg/day for a tramper.

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Date

HUMAN PESTICIDE EXPOSURE ASSESSMENT

California Environmental Protection Agency
Department of Pesticide Regulation
Worker Health and Safety Branch

TRIBUFOS

August 8, 1990
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Introduction

Tribufos is on the list of the first 200 products under the California Birth Defect Prevention Act of 1984 (SB-950). The Department of Pesticide Regulation (DPR) is preparing a risk assessment document for tribufos because the chronic studies have shown possible adverse effects in laboratory animals. Human exposure assessment provides essential information for the risk assessment of pesticides. This human exposure assessment document will be an integral part of the Risk Characterization Document of the DPR for tribufos. It will also serve as a basis for developing mitigation strategies if exposure to tribufos is found to cause excessive risk.

Chemical and Physical Properties

Tribufos (DEF) is the proposed common name for S, S, S-tributyl phosphorotrithioate (CAS No. 78-48-8). It is an organophosphate pesticide that can cause cholinesterase inhibition. Tribufos is marketed in California under the trade names of DEF® and Folex®. Its empirical formula is $(C_4H_9S)_3P{=}O$. Tribufos is a colorless to pale yellow clear liquid with a molecular weight of 314.5, and a vapor pressure of 6.5×10^{-6} mm Hg at 25^{o} C (Talbott and Mosier, 1987). It is soluble in aliphatic and aromatic chlorinated hydrocarbons, and is practically insoluble in water. It is relatively stable in acid and toward heating, but hydrolyzes slowly under alkaline conditions, producing the highly odorous butyl mercaptan. Tribufos can be stored in a freezer and remain relatively stable prior to analysis. The octanol/water partition coefficient for tribufos is 3.31×10^{5} at 25^{o}C (D'Harlingue, 1987). Tribufos contains minute quantities of butyl mercaptan. Butyl mercaptan is a colorless liquid with a strong skunk-like odor. It is highly volatile with a vapor pressure of 35 mm Hg at 20^{o}C (GPC, 1982).

U.S. EPA and California Status

In 1981 the U.S. EPA issued a decision not to initiate the Rebuttable Presumption Against Registration (RPAR) review of tribufos with respect to its neurotoxicity concerns (U.S. EPA, 1981). The U.S. EPA concluded that even though laboratory animal testing indicated potential neurotoxic effects, under actual field use conditions there has not been unreasonable adverse effect to man. Moreover, the U.S. EPA has obtained agreement from the registrants for label changes which specify protective clothing be worn by workers to further reduce the exposure. The U.S. EPA contended in its 1981 decision document that the safety margins for various groups of workers were found adequate.

In California, tribufos was placed into reevaluation on August 9, 1991 since available acute toxicity studies were inadequate to determine whether possible adverse effects were sufficiently mitigated by the signal word and precautionary language on the labels. Additional acute toxicity studies submitted by the registrant indicated that the signal word and precautionary statements on the labels were not adequate to mitigate possible eye and skin irritation hazards. The reevaluation resulted in revising the precautionary statements and changing the toxicity signal word from ‘Warning’ to ‘Danger’.

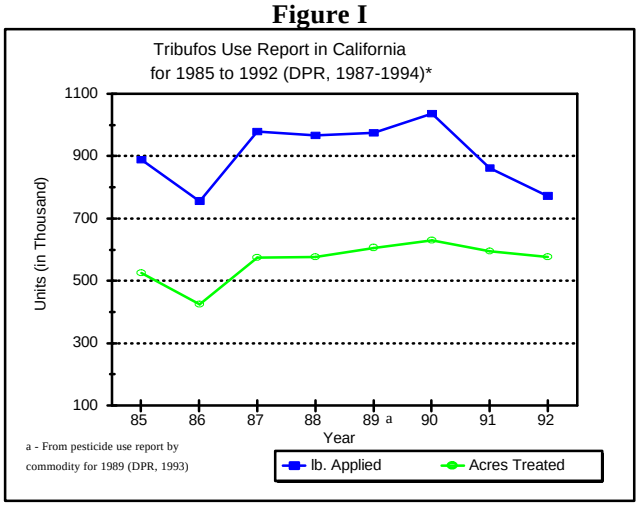
Formulations

To date, there are two tribufos containing products registered in California. They are DEF® 6 Emulsifiable Defoliant and Folex® 6 EC Cotton Defoliant. Both products are emulsifiable concentrates, each containing 70.5 percent of the active ingredient (a.i.) which is equivalent to six pounds (lb.) of tribufos per gallon. Folex was originally formulated with slightly different a.i. (S, S, S-tributyl phosphorotrithioite). Folex with the old formulation was discontinued several years ago. Currently registered Folex contains tribufos as an a.i. and there is no pesticide product registered in California that contains S, S, S-tributyl phosphorotrithioite.

Usage

Tribufos is used exclusively on cotton for defoliation. The recommended application rate is 1.0 to 2.5 pints of the product (0.75 to 1.9 lb. a.i.) per acre. According to label directions, tribufos can be applied as a dilute spray in five to ten gallons of water per acre by air, or ten to 25 gallons of water per acre with ground equipment. It can not be used through any type of irrigation system. Tribufos is applied predominantly by air in California. Under favorable conditions tribufos gives effective defoliation of cotton within four to seven days after application. When continued low temperatures prevail at night (below 60 ^{O}F), complete defoliation may require nine to 14 days.

Figure I



* - Includes both DEF® and Folex®

During 1992, over 6,250 applications were made to 574,170 acres of cotton fields in California using 789,800 lb.
of tribufos (DPR, 1987-1994). The tribufos use trend in California is illustrated in Figure I.

Label Precautions

Both DEF® 6 and Folex® 6EC are toxicity category I products, bearing the signal word Danger on their labels. The precautionary statements on both labels inform users of ingestion and inhalation hazards. The labels also warn users of possible eye and skin injuries. The statement of personal protective equipment (PPE) on the product labels on file (as of March 14, 1995) requires applicators and other handlers to wear the following:

- Coveralls (over long-sleeved shirt and long pants).
- Chemical resistant gloves.
- Chemical resistant footwear plus socks.

- Protective eyewear.
- Chemical resistant headgear.
- Chemical resistant apron when mixing/loading and cleaning equipment.
- MSHA/NIOSH-approved respirator in enclosed areas, or MSHA/NIOSH-approved dust/mist filtering respirator for outdoors.

According to the federal worker protection standards (WPS) for agricultural pesticides [40 CFR 170.240(d)(4-6)], when using closed systems mixing/loading, enclosed cabs, or enclosed cockpits, the PPE requirements for mixer/loaders may be reduced to work clothing (long-sleeved shirt and long pants), chemical resistant apron, and chemical resistant gloves and for applicators may be reduced to work clothing.

Based on the WPS on the labels, restricted reentry interval to treated areas is 24 hours. DEF® 6 label requires a preharvest interval of seven days and a reentry interval of four days for other activities involving human contact with foliage. The Folex® 6 EC label is silent about the preharvest interval and reentry interval for other activities involving human contact with foliage. Tribufos is a restricted material in California. California Code of Regulations (CCR), Title 3, Section 6470 requires a one-half mile buffer zone from residential areas or schools in session (or due to be in session) for all tribufos applications. The regulations also require closed system mixing/loading during tribufos handling. The level of butyl mercaptan in tribufos formulated products must not exceed 0.1 percent according to CCR, Title 3, Section 6361.

Human Illnesses and Injuries

There were a total of 16 illness and injury cases associated with exposure to tribufos and tribufos in combination with other pesticides in California during 1982 to 1991 (DPR, 1994). Of the 16 cases, 11 were systemic, two were eye, and three were respiratory illnesses. Systemic poisoning due to exposure to tribufos and tribufos in combination with other pesticides was positively identified (definite) in four cases. One systemic case was classified as probable, and six others as possible cause of acute poisoning.

In a 1977 report from the California Department of Food and Agriculture, several hundred complaints of human illness were summarized (Maddy, 1977). The illnesses were characterized by wheezing, coughing, nausea, and other discomforts that could be linked to the foul odor of butyl mercaptan, a degradation product of the cotton defoliant. Due to improvements in the manufacturing process of tribufos, a very low odor formulation is available now and has, to some extent, minimized the odorous problem associated with the use of this cotton defoliant. However, this report also emphasized that after the low-odor cotton defoliant has been sprayed on to the field, foul odorous material is generated and may persist for up to 48 hours due to photodegradation and other field conditions (Maddy, 1977). Thus, removing the impurities of the defoliant product alone does not totally eliminate the foul odor.

The are no available clinical reports in this country on human illness due to tribufos exposure. An article entitled "Merphos Poisoning or Mass Panic?" reported a chemical spillage on a ship in Mexico on route to Sydney, Australia (McLeod, 1975). Six hundred and forty-three exposed persons were seen at a local hospital. The most serious problem appeared to be the inhalation toxicity of the butyl mercaptan. It was estimated that the airborne concentration of butyl mercaptan exceeded 0.5 ppm, and in some situations, it had exceeded 10 ppm (ACGIH TLV for butyl mercaptan was 0.5 ppm TWA). Reportedly, there was no cholinesterase inhibition among the tested individuals. In addition to the symptoms usually seen with the exposure of mercaptan, the author emphasized that panic, fear, anxiety, and exhaustion play a major role in exhibiting or intensifying some of the symptoms. Since caustic soda was used for the decontamination process of chemical spillage, more butyl mercaptan was generated, thus, resulting in the continuous supply of foul odorous chemical. The author concluded that there was no significant human illness resulting from organophosphate poisoning in this episode.

Kilgore et al. has conducted medical examination and psychological testing of 14 aerial applicator personnel who were exposed to tribufos (Kilgore, 1984). These volunteers were pilots, flaggers, mixer/loaders and other personnel. Medical examination included a general physical, chest X-ray, EKG, total and RBC cholinesterase,

blood chemistry, and urinalysis. There was no significant medical findings noted in any of the 14 workers. A battery of psychological tests were utilized to measure the neuropsychological functions of the exposed persons. These particular measures were selected because they were considered to be subtle measures of organic brain dysfunction. The test found no significant differences between the pre- and post-exposure scores on any of the psychological measures utilized.

An epidemiological study was conducted by the Department of Health Services on acute health effects associated with the exposure of cotton defoliants (Scarborough, 1989). The study surveyed by phone 460 residents of agricultural communities in the San Joaquin Valley during cotton defoliation. The study found that a positive association exists between the various symptoms and spraying of cotton defoliants for people living or working near a sprayed field. These symptoms included "respiratory allergy", eye irritation, rhinitis, throat irritation, shortness of breath, wheezing, "asthma symptoms", nausea, and diarrhea. In this epidemiological survey it is not known whether the regulation requiring a one-half mile buffer zone between residential areas and the sprayed fields had been enforced during tribufos applications.

Although the very low exposure to tribufos is unlikely to cause a toxicological effect, the presence of an infinitesimal amount of the degradative product (butyl mercaptan) causing foul odor is likely to be associated with the various subjective symptoms and complaints. The TLV - TWA for butyl mercaptan is 0.5 ppm which indicates exposure at this level should not result in untoward acute health effects (ACGIH, 1988). However, the offensive odor which can be detected at 0.001 ppm (Amoore and Hautala, 1983) may have caused the various discomforts and reported illness.

Dermal Irritation/Sensitization

Technical tribufos is a moderate dermal irritant (category III) to rabbits (Sheets and Fuss, 1991). The formulated products are strong dermal and eye irritants (Sheets and Phillips, 1992; Crawford, 1971), which may be due to the inert ingredients. A skin sensitization study with technical grade tribufos found no evidence of dermal sensitization in guinea pigs (Sheets, 1990).

Animal Metabolism

Indirect information indicates differential metabolism (degradation) occurs via various routes of administration. This is likely due to the consequence of kinetics of metabolism. When administered orally tribufos undergoes hydrolysis in the GI tract to produce n-butyl mercaptan, causing late acute effects in test animals without producing neurohistopathological changes. When applied dermally, tribufos does not undergo hydrolysis, thus the late acute effects caused by n-butyl mercaptan are virtually absent. This may explain why topically administered tribufos in large dosages is more effective in producing neurotoxic effects. Other studies also demonstrate that dermally administered DEF was more effective in the inhibition of neurotoxic esterase (NTE) and induction of cytochrome P-450 than orally administered tribufos in adult hens (Abou-Donia, *et al*., 1979, Abou-Donia, 1979, Lapadula, 1984, Abou-Donia, *et al*., 1986).

[^{14}C] tribufos was absorbed rapidly and metabolized extensively by rats when administered orally (gavage) as a single dose of 5 or 100 mg/kg or multiple doses of 5 mg/kg (Kao *et al*., 1991). At least 57 percent of the administered dose was excreted in 24 hours. Approximately 96 percent of the administered dose was excreted in urine and feces within 72 hours. Urinary excretion accounted for 55 to 80 percent of the administered dose. Elimination in feces was 15 to 42 percent of the administered dose. The highest residues in tissues were found in the liver following administration. Of the 18 metabolites detected in urine only one was identified (butyl-gamma-glutamylcysteinylglycine disulfide), accounting for one to four percent of the total dose. No tribufos was found in urine. In feces, the parent compound accounted for 15 to 31 percent of the dose. There was no evidence of bioaccumulation in rats. Metabolism studies in laying hens and lactating goats also showed that tribufos is absorbed rapidly and metabolized extensively (Sahali, 1991; and Hall, 1991).

Dermal Absorption, Route of Exposure, and Toxicity

A recent dermal absorption study of tribufos in rats showed substantial dermal absorption in treated animals (Schroeder, 1992). Adult male Sprague-Dawley rats weighing 200 to 238 grams were used in this study. The back of each animal was shaved and the treated skin site was enclosed with a rubber ring. The animals were placed in individual metabolism cages to allow the separate collection of urine and feces during the study. Rats were treated at three dose levels of 1.93, 12.4, and 100 ug/cm^2 . The dosing solution was tribufos-1-^{14}C (98.9% purity) mixed in distilled water with DEF 6 blank formulation and nonlabeled tribufos as needed. The treated skin site was protected with a non occlusive cover made of a Teflon-laminated filter and a carbon-impregnated material. Rats were exposed for 10 hours or until sacrificed whichever came first. A group of four rats from each dose level were sacrificed at 1, 4, 10, and 168 hours. All sampling media such as non occlusive cover, ring wash, treated skin site, cage wash, carcass, urine, feces, and blood were collected for analysis. Recoveries of the administered doses ranged from 89 to 106 percent. Tribufos was apparently readily absorbed because the skin residues for all dose levels and exposure periods were similar. Dermal absorption was calculated as the sum of percent dose in urine and feces at asymptote and percent dose recovered in, carcass, cage wash, and blood. The dermal absorption values after correction for recoveries were 47.5, 47.9, and 33.9 percent for the low, medium, and high doses, respectively. The dermal absorption of 47.5 percent for the low dose will be used in worker exposure estimates because this dose is closer to the level of exposure experienced by workers handling tribufos or tribufos treated cotton.

Dislodgeable Foliar Residue

A cotton boll residue study was conducted in conjunction with a cotton harvester exposure study (Eberhart and Ellisor, 1993). The harvester exposure portion of the study is discussed in the Worker Exposure section of this document. The residue portion of the study was conducted as follows. Cotton boll residue samples were taken at two locations (California and Mississippi). Two separate residue trials were conducted at each location. In California, cotton boll samples (50 grams each) were collected from the fields harvested during the worker exposure monitoring portion of the study. The fields were treated with DEF 6 at maximum label rate (2.5 pints/acre) either aerially or by ground equipment. Triplicate samples were taken from aerially-treated field prior to the application and at 0, 1, 2, 4, 7-13, 15, and 17 days post application. Triplicate samples were also taken
from ground-treated fields at the same intervals and 16, 18, and 20 days post application. The cotton boll samples were placed in a container with 200 mL of Nekal/water solution and shaken for 20 minutes. The solution was decanted into a pre-labeled container. Control and field fortified samples were also taken. The study was performed according to the U.S. EPA's Good Laboratory Practice (GLP) standards. The study combined the cotton boll residue data from California and Mississippi to predict residue degradation. Residue degradation is highly environmentally dependent. We prefer to use California data when readily available. The predicted residue levels in Table 1 were derived from log-linear regression analysis of observed residues in California over time.

In a defoliated cotton field, cotton bolls could be the primary source of pesticide exposure for cotton harvesting crews entering the treated field. Cotton boll residues are not necessarily dislodgeable residues as conceived with other crops, but can be used for an indirect estimate of exposure of workers involved in harvesting.

Table 1
Tribufos Residue Levels in Cotton Bolls in California (ug/g)

Post Application	Observed (average)		Best Fit Curve	
Days	*ground*	*aerial*	*ground*	*aerial*
0	3.91	2.36	1.81	1.95
1	2.59	2.62	1.49	1.50
2	1.18	1.39	0.85	0.69
4	0.57	1.39	0.85	0.69
7	0.38	0.23	0.47	0.32
8	0.40	0.14	0.38	0.25
9	0.17	0.15	0.32	0.19
10	0.14	0.14	0.26	0.15
11	0.14	0.02	0.22	0.11
12	0.15	0.05	0.18	0.09
13	0.18	0.06	0.15	0.07
15	0.09	0.05	0.10	0.04
16	0.06	n.c.	0.08	0.03
17	0.09	0.14	0.07	0.02
18	0.10	n.c.	0.06	0.02
20	0.06	n.c.	0.04	0.01
r^2			0.88	0.74
slope			-0.19	-0.26
$t_{1/2}$ (days)			4.67	3.77

* - n.c. - No samples were collected.

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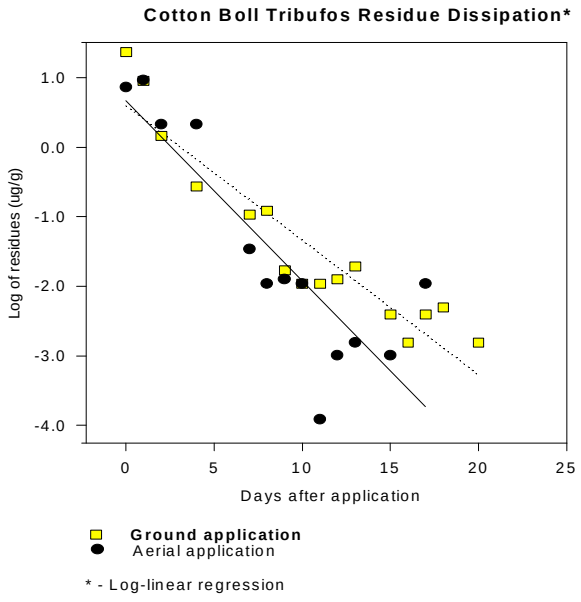
Figure 2



Worker Exposure

Exposure of Handlers:

1. Handler Exposure Study (Peoples *et al.*, 1981)
   Two aerial applicator firms cooperated in this study. Monitoring included inhalation and dermal exposure of mixer/loaders, pilots, and flaggers during the applications of DEF® and Folex® in the San Joaquin Valley of California in 1979. Closed system transfer was used during this study by both firms. However, the containers were rinsed by hand, and the rinse water was hand-poured into the mix tank by mixer/loaders of one firm (firm #2). The application rate of the defoliants was 0.22 and 0.25 gallon per acre (approximately one pound a.i. per acre), and the volume of spray was 10 gallons of water per acre.

   Mixer/loaders wore rubber boots, socks, a shirt, pants, and a washable cap. Clean long-sleeved and long-legged coveralls were required daily. The mixer/loaders wore neoprene gloves when hooking up, loading, and washing the aircraft. They removed the gloves between mixing/loading operations and while cleaning the nozzles. The pilots wore shoes and socks, a helmet, and clean long-sleeved shirts and long-legged cloth pants, which were changed daily. They are expected to wear rubber gloves when adjusting spray nozzles. Some pilots did not wear gloves when adjusting spray nozzles. The flaggers wore clean coveralls (with long sleeves and legs) and washable caps.

   Inhalation exposure was monitored by placing a Dupont Constant Flow Sampler P-4000 pump on each worker with the air intake placed in the breathing zone. Air sampling tubes containing Amberlite XAD-4 resin were used as the sampling media. Dermal exposure was measured with patches made of an outer layer of cloth and an inner layer of gauze taped together. The outer layer represented the protective factor of clothing and the inner layer represented the skin surface. Patches were taped to the clothing and skin areas. The residues on the head and neck surfaces were measured as the sum of the inner and outer patches. Hand exposure was monitored at the end of the workday by rinsing the hand with ethyl alcohol, after removal of gloves when applicable.

Table 2
Estimates of Absorbed Daily Dosage of Workers Exposed
to Tribufos During Aerial Application

Work Activity (n)	Exposure[a] (ug/person/day)					(ug/kg/day)
	Head/neck	Body	Hand	Total dermal	Inhalation	ADD[a, b]
Mixer/Loader (10)	1965	2169	5617	11398	257	74.4 ± 1.7
Pilot (11)	518	1790	4699	8354	124	53.1 ± 1.5
Flagger (11)	1266	202	175	1897	313	15.2 ± 2.6

a - Geometric mean (log-normally distributed) and seven-hour workday.
b - Dermal absorption of 47.5 percent, inhalation rate of 14 L/minute and respiratory uptake of 50 percent, body weight of 75.9 kg, and workday of 7 hours.
(n) - Number of observations.
For pilots wearing work clothing (long-sleeved shirt, long pants, socks, and shoes). Mixer/loaders wearing work clothing, gloves, and apron and used a closed mixing/loading system. Flaggers wearing work clothing, coveralls, gloves, hat, and a dust mask. Coveralls, gloves, hat, and apron, each providing 90 percent dermal protection to the covered areas. Dust mask providing 50 percent respiratory protection.

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The estimates of exposure were normalized to seven hours. The monitoring days were selected so that at least seven hours of continuous applications were expected but the monitoring period varied from a few hours to 7 hours. The actual application time was 6 to 12 hours daily. Gas chromatography (GC) was used for the analysis of tribufos in collected samples following solvent extraction. Table 2 shows estimated dermal and inhalation exposure of workers to tribufos during aerial application.

2. Handler Exposure Study (Eberhart, 1993)

Dermal exposure, inhalation exposure, and cholinesterase activities of handlers were monitored during tribufos application to cotton at two locations in San Joaquin Valley, California and one location in Glendora, Mississippi.

Four commercial applicator crews were monitored in California. At the first location, two crews, each consisting of a pilot, a mixer/loader, and two flaggers, applied DEF® 6 by air at 2.5 pints (1.9 lb. a.i.)/acre and 1.5 pt (1.1 lb. a.i.)/acre. A total 1710 gallons of DEF® 6 was applied aerially to 6705 acres of cotton during the monitoring. At the second location, two crews, each consisting of one mixer/loader and one applicator applied DEF® 6 by ground rig boom sprayers equipped with closed cabs and air conditioning at 2.5 pints./acre. A total of 165 gal. of DEF® 6 were applied to 531 acres of cotton. Closed mixing/loading systems were used at both locations in California. In Mississippi, only mixer/loaders of two aerial crews were monitored during open mixing-loading operations.

Dermal and inhalation exposure of workers was monitored during four one-half day (approx. 4 hours) periods. Each worker wore a dosimeter garment (a long-sleeved tee-shirt and a pair of tights) under a single layer of coveralls (as work clothing), and a hat. All mixer/loaders wore additional protective clothing consisting of nitrile gloves, goggles, and rubber boots. Gauze patches were attached to the coveralls at the chest, back, and front of the cap to estimate exposure to uncovered areas such as neck, face and head. Hand exposure was measured by taking ethanol hand washes at the end of each monitoring period. An air sampling pump connected to an OVS-2 tube (glass fiber filter backed by two sections of XAD-2 resin) was attached to the belt of each worker to monitor air residues in workers' breathing area. All collected samples were placed on dry ice and shipped via over-night delivery to the laboratory. RBC and plasma cholinesterase activities of three to five workers in each job category were monitored weekly for a 3-4 week period during the rest of cotton defoliation season in California. The workers participating in cholinesterase monitoring were not necessarily all participants of the exposure monitoring portion of the study. The rate of applications varied during cholinesterase monitoring. Blood samples were analyzed as soon as possible, mostly within 48 hours of collection.

Control and field fortified samples were prepared on each day of monitoring. The study was performed according to the U.S. EPA's GLP standards. Blood cholinesterase analyses were performed in a manner consistent with GLP. All exposure results were corrected for field recoveries. Most field recoveries were over 90 percent. Head and neck surface area as described in the U.S. EPA Subdivision U Guidelines were used in calculating dermal exposure to those exposed areas. The exposure values were reported as ug/lb. handled, ug/hour, and ug/replicate.

Table 3
Mixer/loader, Applicator, and Flagger Exposure to Tribufos During
DEF 6 Application to Cotton in San Joaquin Valley, California

Job Category (n)	Exposure[a] (ug/person/hour) Head/neck	Body	Hands	Total Dermal	Inhalation	ADD[a, b] ug/kg/day
Aerial Application						
M/L (8)	182.2	199.8	181.9	629.6	9.5	28.1 ± 2.2
Pilot (8)	67.8	100.2	542.7	748.1	5.0	33.2 ± 2.8
Flagger (16)	520.5	43.6	78.1	657.7	11.6	29.6 ± 2.5
Ground Application						
M/L (8)	243.0	252.2	434.0	1075.8	8.0	54.3 ± 2.0
Applicator (8)[c]	4.6	58.8	19.6	86.8	1.4	4.4 ± 1.6

a - Geometric mean (log-normally distributed).
b - Dermal absorption of 47.5 percent, inhalation rate of 14 L/minute and respiratory uptake of 50 percent, body weight of 75.9 kg, and workday of 7 hours for aerial crews and 8 hours for ground crews (± standard deviation of geometric mean).
c - In enclosed cab wearing work clothing.
(n) - Number of observations.
For pilots wearing work clothing (long-sleeved shirt, long pants, socks, and shoes). Mixer/loaders wearing work clothing, gloves, and apron and used a closed mixing/loading system. Flaggers wearing work clothing, coveralls, gloves, hat, and a dust mask. Coveralls, gloves, hat, and apron, each providing 90 percent dermal protection to the covered area. Dust mask providing 50 percent respiratory protection.

Formoli, WH&S, 1993

3. Handler Exposure Study (Lotti, 1983):
Lotti, et al. reported a worker exposure study consisting of seven workers during aerial application of DEF in cotton fields. The dermal exposure data generated from this study was neither meant for, nor appropriate, to the whole body dose extrapolation. The primary objective of this study was to determine whether the measurement of neurotoxic esterase (NTE) inhibition was useful in monitoring workers exposed to organophosphates that can cause delayed neurotoxicity.

In addition to dermal exposure and air monitoring, a series of tests to monitor peripheral nerve function and nerve enzyme activity were conducted. These tests are collectively called electrophysiology and biochemical measurements, emphasizing the subclinical aspects of worker exposure. By definition, subclinical effects denote the subtle changes of enzyme markers or electorphysiological changes of nerve function which by themselves do not lead to demonstrable clinical symptoms.

Prior to conducting these tests, workers were exposed to the defoliants for a lengthy period (averaging 27 days), thus, the test results are considered reliable as indicators of subtle effects following season long exposure. The exposure did not result in any detectable clinical effect on the peripheral nervous system, nor was there any cholinesterase inhibition detected in any of the exposed workers. The lymphocyte NTE activity was significantly inhibited in most of the exposed workers. This inhibition appears to be correlated to the length of exposure and did rebound approaching normal values three weeks after the exposure was terminated. The significance of the NTE inhibition and the potential of developing neuropathy is not known. However, the measurement of NTE during the exposure to organophosphate may be a useful biomarker and serve as a warning signal for overexposure.

Exposure of Cotton Harvesters:

1. Cotton Harvester Exposure Study (Roberts and Smith, 1980; Cox et al., 1980 [a, b, c])
   Mechanical cotton picker operators' dermal and respiratory exposure to tribufos was studied in California at four locations. Closed cab, two-row type mechanical cotton pickers were used to harvest cotton. Respiratory samples were collected inside the cab at the operator's breathing zone, using air sampling pumps calibrated at 1.0 liter per minute. Dermal exposure was monitored by attaching gauze pads with polyethylene backing to the front thigh clothing areas of the operators.

   Only one pad was used for each operator to monitor dermal exposure. The weather conditions prior to and during this study were unusual (low temperature, high humidity, rain) for the season and not typical of California. The study authors concluded that the unusual weather conditions could contribute to reduced exposure levels. In the presence of more reliable data (Eberhart and Ellisor, 1993), the information from this study may not be appropriate for estimating tribufos exposure to cotton picker operators.

2. Cotton Harvesters Exposure Study (Eberhart and Ellisor, 1993)
   A more recent study monitored dermal exposure, inhalation exposure, and cholinesterase activities of five cotton harvesting crews in San Joaquin Valley, California. DEF® 6 was applied to the cotton fields at the label rate of 1.9 lb. a.i./acre either by aerial or ground equipment (Eberhart, 1993).

   All crews used mechanical harvesters to harvest cotton. Three crews used mechanical module builders to compact the harvested cotton. The other two crews compacted the harvested cotton by the old fashion method

Table 4
Mean (arithmetic) Dermal and Inhalation Exposure of Workers
to Tribufos During Cotton Harvesting (Eberhart and Ellisor, 1993)

Job Category	Dermal Exposure (ug/person/hr)[a] exposed	covered	Hands	Total	Inhalation[a] (ug/person/hr)	ADD[a,b] (ug/kg/day)
15 Days After Aerial Application:						
Picker Operator	19.3	33.8	32.1	85.2	4.0	4.5 ± 1.4
Module Builder Operator	10.6	61.2	7.7	79.4	4.2	4.2 ± 1.6
Raker	22.6	74.9	32.2	129.8	4.7	6.8 ± 2.2
17 days After Aerial Application:						
Picker Operator	12.8	60.6	63.8	137.2	1.9	7.0 ± 1.6
Module Builder Operator	4.0	9.3	7.3	20.6	1.7	1.1 ± 0.1
Raker	9.9	31.7	11.8	53.4	1.1	2.7 ± 1.1
20 Days After Ground Application:						
Picker Operator	21.6	74.4	116.9	212.9	1.8	10.7 ± 1.5
Tramper	15.0	122.6	108.7	246.3	4.0	12.5 ± 8.9
Raker	6.6	58.4	38.2	103.3	2.5	5.3 ± 2.2

a - All data are arithmetic means (data mostly normally distributed).
b - Based on dermal absorption of 47.5 percent, inhalation rate of 14 L/minute and respiratory uptake of 50 percent, body weight of 75.9 kg, and workday of 8 hours, (± standard deviation).
Clothing : A long-sleeved shirt, long pants, socks, and shoes.

Formoli, WH&S, 1993

of physical tramping. Workers of each crew were divided into three job categories as harvesters (picker operators), compactors (module builder operators or trampers), and rackers who also picked loose cotton on the ground. Crews with mechanical module builders entered the cotton fields 15 and 17 days after a single aerial application. Crews with trampers entered cotton fields 20 days after a single ground application. Dermal and inhalation exposure of workers was monitored during two 4-hour periods. Each worker wore the dosimeter garments (a long-sleeved tee-shirt and a pair of tights) under a single layer of coveralls (as work clothing). A hat was also worn that held a gauze patch. Gauze patches were attached to the coveralls at the chest, back, and front of the hat to estimate exposure to uncovered areas such as neck, face and head. Hand exposure was measured by taking ethanol hand washes at the end of each monitoring period. An air sampling pump connected to an OVS-2 tube in worker's breathing zone (glass fiber filter backed by two sections of XAD-2 resin) was attached to the belt of each worker to monitor inhalation exposure. RBC and plasma cholinesterase activities of five workers in each job category were monitored weekly for a 5-6 week period during the rest of cotton harvesting season. All exposure results were corrected for field recoveries. The participating workers spend 31 to 35 days per year performing cotton harvesting activities. Average body weight for workers (nine males and seven females) was 75 $\pm$13 kg. Table 4 shows the estimates of exposure for these workers.

Mean five-week plasma and erythrocyte cholinesterase values for each worker were within 88 to 107 percent of his/her baselines. The two lowest erythrocyte cholinesterase activities observed were both 77 percent of the baseline for two module builder operators on the fifth week of monitoring. The next lowest erythrocyte cholinesterase observed was at least 83 percent of the baseline. The lowest plasma cholinesterase activity observed was 79 percent of the baseline for another module builder operator on the fourth week of monitoring and raised to 85 percent of the baseline on the fifth week of monitoring. While cholinesterase activity is a general indicator of exposure, it is not a gauge to specifically quantify exposure.

To estimate the dermal exposure of harvesters entering treated fields seven days after tribufos application, a dermal transfer factor was calculated from the estimate of dermal exposure of workers in Table 4 and the level of residues in cotton bolls (Table 1) on the day worker exposure was measured. Table 5 shows the estimate of exposure of harvesters entering treated fields immediately after the expiration of the preharvest interval. The indirect estimation of field workers' exposure from the level of dislodgeable foliar residues is an acceptable method in exposure assessment; however, cotton boll residues are not foliar dislodgeable residues as conceived with other crops. Cotton boll residues are probably distributed throughout the boll while foliar dislodgeable residues are only surface residues.

Table 5
Tribufos Estimated Dermal Transfer Factors and ADDs for Workers
Involved in Cotton Harvesting Following Ground or Aerial Application

Days After Application	Dermal Exposure Observed ug/hr	Cotton Boll Residue Predicted ug/g	Dermal Transfer Factor g/hr	Dermal Exposure Calculated ug/hr	ADD[a] Calculated ug/kg/day
Picker Operator:					
15	85.2	0.04	2131		
17	137.2	0.02	6859		
20	212.9	0.04	5323		
Average			4771		
7 (Ground)		0.47	4771	2242	112.2
7 (Aerial)		0.32	4771	1527	76.4
Module Builder Operator:					
15	79.4	0.04	1985		
17	20.6	0.02	1029		
Average			1507		
7 (Ground)		0.47	1507	708	35.4
7 (Aerial)		0.32	1507	482	24.1
Raker:					
15	129.8	0.04	3245		
17	53.4	0.02	2669		
20	103.3	0.04	2582		
Average			2832		
7 (Ground)		0.47	2832	1331	66.6
7 (Ground)		0.32	2832	906	45.4
Tramper:					
20	246.3	0.04	6157		
7 (Ground)		0.47	6157	2894	144.9
7 (Aerial)		0.32	6157	1970	98.6

a - Assuming dermal absorption of 47.5 percent, body weight of 75.9 kg, 8-hour workday, negligible inhalation exposure, and workers wearing work clothing.

Formoli, WH&S, 1993

Cotton defoliation is a seasonal activity in California. It starts in early September (Southern San Joaquin Valley) and ends in mid October (Central San Joaquin Valley), depending on the weather conditions and crop maturity (Vargas, 1993; Wright, 1993). According to the product label, tribufos effectiveness is limited by temperature. The climatic conditions favorable for effective use of tribufos normally occur during the first two to three weeks of the defoliation season (Wright, 1993). A worker may handle tribufos an average of four to five workdays during the season (Haskell, 1993). Three weeks (21 workdays) in a season may be the maximum. The seasonal average daily dosage (SADD) and annual average daily dosage (AADD) in Table 6 are based on three weeks of exposure in a 45-day season and in a 365-day year respectively for both tribufos handlers and cotton harvesting crews.

Table 6 is a summary of the estimates of exposure for tribufos handlers and cotton harvesting crews. The estimates of ADD for mixer/loaders and pilots in Table 6 are based on Eberhart, 1993 exposure study since the closed system technology and the work habits in this study are more representative of the current use of tribufos. During Peoples, *et al.*, 1981 study, protective clothing were worn irregularly. Some pilots adjusted

the nozzles without gloves. Mixer/loaders removed their gloves during handling and the mixing/loading procedure of one crew involved hand pouring. The estimates of ADD for flaggers is based on weighted average of both studies.

Table 6
Estimated ADD and AADD for Tribufos Handlers, and Field workers

Job Category	ADD ug/kg/day	SADD ug/kg/day	AADD ug/kg/day
During application:			
Mixer/loader (aerial)	28.1	13.1	1.6
Pilot	33.2	15.5	1.9
Flagger	23.7*	11.1	1.4
Mixer/loader (ground)	54.3	25.3	3.1
Applicator (ground)	4.4	2.1	0.3
Seven days postapplication:			
Picker Operator	94.3	44.0	5.4
Module Builder Operator	29.8	13.9	1.7
Rake	56.0	26.1	3.2
Tramper	121.8	56.8	7.0

* - Weighted average of ADDs in Tables 2 and 3 (Peoples *et al*., 1981 and Eberhart, 1993)
A 21-workday tribufos application or harvesting in a 45-day season or in a 365-day year was assumed.

Formoli, WH&S, 1993

Exposure of Non-Applicator Personnel and Crews:

1. Drift Study (Lowrimore *et al*., 1985)
   The primary objectives of this study were to measure the air drift capacity of tribufos, and to monitor the potential dermal exposure through measuring the deposition of tribufos on denim cloth under field study conditions. This study was conducted in Pickins, Arkansas, on six cotton fields ranging from 46 to 70 acres. The results indicated that at one kilometer (0.6 mile) downwind and within 0 - 60 minutes after spraying, the amount of tribufos found on denim ranged from 0.03 to 0.34 ug/cm^2, with the majority of the samples below 0.1 ug/cm^2. Table 7 shows the average levels of tribufos in denim patches and air monitors. There was no attempt at odor surveillance in this study.

Table 7
Exposure of Non-Applicator Personnel and Crews to Tribufos (Lowrimore *et al*., 1985)

Distance	Within 0 to 60 minutes After Application		Within 60 to 180 minutes After Application	
Downwind (meters)	Denim Patch (ug/100 cm^2)	Air (ug/M^3)	Denim Patch (ug/100 cm^2)	Air (ug/M^3)
10	993	10	9	2
50	119	8	4	1
100	46	6	5	1
1000	10	not reported	4	not reported

Wang, WH&S, 1990

2. Drift Study (Oshima *et al*., 1980)
   The California Department of Food and Agriculture (CDFA) conducted this study in September and October of 1979 in Fresno and Merced Counties to monitor tribufos downwind drift during and following aerial applications. The CDFA also monitored ambient air of residential areas (Mendota and Dos Palos) for tribufos. Tribufos levels in the air during applications ranged from 14.5 ug/m^3 to 0.95 ug/m^3 between 30 and 400 meters downwind from the field. Tribufos levels several hours after the termination of applications were close to the background (0.6 ug/m^3) that was detected prior to the field applications. No tribufos was detected in the ambient air samples of residential areas.

3. Drift Study (Seiber *et al*., 1983)
   This study monitored ambient air levels of several cotton harvest aid chemicals including tribufos during commercial aerial applications in the San Joaquin Valley. Air samples collected during and following the application of tribufos were analyzed for tribufos, n-butyl mercaptan, and dibutyl disulfite. Samples collected during application of tribufos at 50 meters distance contained 1.19 , 0.002, and 0.015 ug/m^3 of tribufos, dibutyl disulfite, and butyl mercaptan, respectively. Residues decreased to 0.45, 0.0005, and 0.004 ug/m^3 of tribufos, dibutyl disulfite, and butyl mercaptan, respectively, 24 hours following application.

4. Ambient Air Monitoring Study (Seiber *et al.*, 1988)-
   Ambient air monitoring for tribufos was conducted during September and October 1987 at four residential areas in Fresno County (Tranquility, San Joaquin, Five Points, and Huron) and two urban background locations in cities of Fresno and Bakersfield. The four locations were in the proximity of cotton fields ranging from 10 to 400 meters from the edges of the fields. High volume air samplers equipped with XAD-4 resin sampling tubes were used. A total of 164 field samples were collected. No tribufos was detected (MDL = 0.001 ug/m^3) at urban sites except for two days in Bakersfield and four days in Fresno where the tribufos concentrations were above the MDL. Tribufos levels at four residential areas ranged from a minimum of below the MDL to a maximum of 0.34 ug/m^3.

The data generated from these studies indicate that the exposure to work crews in adjacent and near-by fields is minimal compared to that of workers involved in handling tribufos or harvesting tribufos treated cotton. The potential exposure of the general population to tribufos is assessed in a separate document (Formoli, 1994) prepared under the Assembly Bill 1807.

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